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Differential observations in spectroscopic measurements using electron beam ion traps

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Abstract
In many atomic physics experiments in an electron beam ion trap, the technical system is used to provide a stationary environment for a cloud of highly charged ions which are then probed for properties such as excitation energies or magnetic sublevel population (via the polarization of the emitted light). However, there are also observations in which electron beam properties or ion trapping conditions are systematically varied to obtain atomic properties as well as measurements of the changes over time of the stored ions, their atomic states and their ensemble properties. Examples of such measurements that require a variation of the experiment parameters (hence 'differential' observations) are discussed.

1. Introduction

Ion traps are marvellous tools for a great variety of atomic physics experiments. Probably, the majority of such traps are being used for singly charged ions that can be interrogated by laser light. Electrostatic (Kingdon) and magnetic (Penning) ion traps have also been used to store multiply charged ions, as have radiofrequency (Paul) traps. Apparently more suitable to the task is, however, the electron beam ion trap [1] which can be seen as a Penning trap with a built-in electron beam that helps in producing highly charged ions inside the trap and—via space charge effects—with the trapping of the ion cloud. (An electron beam can trap an ion cloud even without an external magnetic field [2, 3].) The spatial symmetry impressed on the collision system by the unidirectional fast electron beam can be exploited in polarization studies that reveal non-statistical population of magnetic sublevels [4–6], and the same polarization effects have to be taken into account when tracking changes of the excitation cross sections of spin–orbit mixed levels along isoelectronic sequences [7].

If one switches off the electron beam after making a cloud of highly charged ions, the positively charged ion cloud significantly expands, but remains confined in the Penning trap [8]. When the electron beam is switched on again, the old ion cloud does not contract to its former small size, but a new one is produced along the electron beam. Evidence for the incomplete contraction of the old ion cloud is seen in atomic lifetime measurements in which the electron beam is switched off so that the excitation ends and radiative decays can be observed. If all ions returned to the same cloud size as before when switching the electron beam back on, the signal recovery should be exponential with a similar time constant as the radiative decay, but in fact (see [9, 10]) it is markedly slower and follows a more complex curve. (With a deeper ion trap used in recent measurements elsewhere, the effect is less pronounced, but statistically even clearer.) Evidently a new sample of ions in the right charge state has to be bred first.

The electron density in the electron beam is much higher than the ion density in the same volume, although (because of space charge compensation) the ion density can be much higher than without the beam. A plasma with a net negative charge is thus produced on the axis (defined by the electron beam) of the previously produced positive ion cloud. Evidently, these are non-neutral plasmas, and a plethora of plasma dynamical experiments should be possible with them (see [11] and the conference proceedings that paper was part of).

However, such collision system symmetries or plasma dynamics are not what I want to discuss here, but rather the information that can be gained by differential observations when changing the running conditions of an electron beam ion trap, in particular the energy and/or the current of the electron beam. This goes beyond the more typical experiments performed at practically all EBITs in which, for example, the electron beam energy is varied through a range of values; mapping the energy-dispersed x-ray signal as a function of the electron beam energy [12] makes it possible to find the position of dielectronic recombination (DR) resonances and to learn about radiative recombination (RR) processes, see also reviews in [13]. Instead, I discuss atomic properties that can be measured or at least inferred from an electron beam modulation experiment and which may influence the internal dynamics of a plasma (such as the charge state distribution; the motion of the plasma cloud under the influence of external fields is yet another matter). Such entities are atomic lifetimes, the identification of charge states in atomic spectra and the response of charge state distributions to changes in external parameters because of the presence and the finite atomic lifetimes of metastable levels.

The electron beam in an EBIT serves to ionize itinerant atoms of the residual gas in the ultrahigh vacuum (UHV) vessel or of a ballistically injected gas plume (still at UHV particle densities) so that they can be trapped by the electric potential difference of collinear drift tubes and axially confined by a strong (several Tesla) magnetic field. Instead of starting from neutral atoms, one can inject ions in low charge states from an external ion source, for example a metal vapour vacuum arc (MeVVA) source, that drift along the magnetic field into the trap volume. The ions will mostly be confined to the volume of the electron beam, and there they can be hit again and again. Further ionization stops when the next step would require to overcome an ionization potential higher than the electron beam energy (a seeming exception is discussed below). The actual charge state balance in the ion cloud depends on ionization and recombination processes with energetic beam electrons or by charge exchange (CX) with neutral particles of the residual gas. Under clean enough vacuum conditions, a 200 keV electron beam can fully ionize uranium and reach U^{92+} ions [14]. Hence ions of all charge states of all elements can be produced. The highest charge state produced can be selected by external parameters. This tool can be exploited for the separation of complex spectra of consecutive charge states of a given element (section 2).

Most ions will be in their ground state because the cross sections for excitation are small for collisions with energetic electrons, and the electron density in the beam is rather low for any laboratory plasma, in the range near $n_e = 10^{11}$ cm^{-3}. The collision rates are lower than most radiative rates, and the ions usually have time to return to the ground state or to a low-lying metastable level before they are hit again. Consequently, the EBIT spectra are dominated by the decays of levels that can be excited from the ground state directly. This is in stark contrast to the interaction of fast ions with a thin foil target (solid state density) [15]. Some other levels may be populated as a consequence of recombination or CX. Drastic changes in the level population after CX have been observed at very low collision energies (sub-keV to eV); such low-energy CX collisions of highly charged ions with neutral gas atoms are discussed as the source of x-ray emission from comets [16]. CX is of interest beyond the cometary context; various surprising observations have instigated research into slow collisions between very highly charged ions and neutral atoms or molecules. Decades ago, such research employed recoil ions produced by distant collisions of fast primary heavy ions on a gas target; an EBIT meanwhile offers to widen the parameter space that is accessible to such collision studies [17–19].

An important point to recognize about the EBIT is that all excitation stops when the electron beam is switched off. CX processes continue, however, and radiative decays take place, but ionization and collisional excitation by swift electrons end. A steady-state dynamic balance thus is replaced by non-equilibrium conditions. This offers a window into measuring time-dependent processes such as the radiative decay of metastable levels (section 3).

It is not always necessary or appropriate to switch the electron beam off completely. It is also possible to vary the electron beam energy and current in order to simulate a Maxwellian electron energy distribution [20] or to change in a controlled manner between two settings and the associated steady-state conditions. Out of the multitude of possible experiments, I will present one in which atomic properties visibly influence the transition to a new steady-state equilibrium (section 4).

2. Charge state analysis of spectra

A persistent problem in spectral analysis is the determination not only of the element that emits a given spectral feature, but also the charge state, before it makes sense to assign a transition. The historic path has been the intercomparison of spectra recorded of a flame, an arc or a spark, but this covers only the first few ionization stages. If one changes the parameters of a spark discharge, one changes the charge state distribution, but has no information on how much. The same holds true for fast ion beams in beam–foil spectroscopy or laser-produced plasmas. In contrast to this, the electron beam energy in an electron beam ion trap can be adjusted in a very controlled way, steering for a defined maximum charge state. Certain problems arise from the energy width of the beam, which is usually between 30 and 50 eV, space charge effects and contact potentials. For relatively low charge states, the ionization potentials may differ by less than the energy width of the electron beam. However, if done carefully, spectra recorded at selected electron beam energy may differ by the contributions of a single charge state. The difference spectra then may represent the spectral information from this single charge state spectrum.

This technique has been demonstrated at Livermore especially in pursuit of E1-forbidden optical lines [21] and for the extreme ultraviolet (EUV) spectra of Fe^{7+} to Fe^{9+}, Ar^{8+} to Ar^{15+} and S^{6+} to S^{13+} [22–24]. At the Tokyo EBIT laboratory, in addition to the large EBIT already operating,

a small EBIT has been built for applications in astrophysics. With this device, the next higher charge states of Fe have recently been studied in the same way [25, 26]. The Japanese group used a flat-field EUV spectrometer basically similar to the Livermore equipment and also a CCD camera. In this way, the difference spectra become available by channel-by-channel subtraction of spectral recordings that each has about 1000–1300 data channels spanning a spectral range from, say, 3 to 10 nm or from 8 to 20 nm. In such a range, some 30–100 spectral lines are seen, both in EBIT and in spectra recorded on board of spacecraft such as *Chandra* or *XMM-Newton* pointing at astrophysical objects in outer space. Of these, many arise from Fe ions (because of the high cosmic abundance of Fe) and some 2–15 spectral lines can typically be identified with a specific charge state.

A group from Dublin working at the NIST EBIT has studied EUV spectra of many charge state ions of xenon this way [27], the line identification in their moderate-resolution spectra benefiting from high precision wavelength measurements in earlier work at other light sources. The Heidelberg EBIT group, having done corresponding work on Fe as well, has recently also produced maps of such spectra for xenon [28]. The recent surge in studies of EUV spectra of multiply charged Xe ions (including this and other work at EBITs) reflects the physicists' preference for a Xe-based EUV light source with bright emission at a wavelength of 13.5 nm that might be used for EUV lithography, whereas the semiconductor industry has agreed on a roadmap that intends to use Zn as the element of choice.

Magnetically confined fusion plasmas are among the contenders for future fusion power reactors. In the latest project (ITER), the surfaces expected to have to bear particularly high particle flux and radiative heat loads are planned to be made of or at least coated with tungsten (nuclear charge $Z = 74$). It is expected that sputtered tungsten atoms will drift into the fusion plasma and contaminate it. The atoms will be ionized corresponding to the electron energies encountered in the plasma and therefore they can serve as probes to their environment via an investigation of the radiation they emit. However, the many ionization stages of a heavy element such as tungsten relate to as many spectra, many of them complex. In order to disentangle them, systematic studies of tungsten spectra recorded at many electron beam energy settings have been employed at several electron beam ion traps [29–34], answering to the explicit needs of the magnetic fusion community [35–37].

3. Atomic lifetime measurements

The highest charge state reached in an EBIT is limited by the electron beam energy. In two-electron ions (He-like spectrum), the lowest triplet level, 1s2s ^{3}S$_1$, has a much higher excitation energy than the ionization energy of the next lower charge state, the three-electron ion (Li-like spectrum), and therefore it can be excited rather efficiently. The magnetic dipole (M1) decay of this level is described by a purely relativistic operator. The line ('z' in plasma physics parlance) is prominent in many low-density plasmas and it plays a role in plasma diagnostics. The line was first recognized in solar x-ray spectra [38] against a theoretical verdict by Breit and Teller [39] that the transition could not happen by a single photon decay. By now, the rate of this transition has been measured from He ($Z = 2$) to Xe ($Z = 54$), finding an upper level lifetime that varies by 15 orders of magnitude over this range. Beam–foil spectroscopy has targeted this transition with measurements from $Z = 16$ upwards. In the higher-Z part of the range (picosecond lifetimes), the results scatter by up to 10% around the predicted trend. Near $Z = 18$ (Ar), however, the experimental error of the many-hundred nanosecond lifetimes initially was deceptively low, while the actual results differed massively from expectation [40]. Certain oversights in the early experiments have been recognized meanwhile and the discrepancies removed. However, it was an important step when work at the TSR heavy-ion storage ring at Heidelberg and at the Livermore EBIT not only extended the range of lifetimes covered to the millisecond and eventually second range, but did so with an accuracy of 1% and better (see general discussions in [41, 42]).

The first EBIT experiment of the series was on Ne^{8+} [43]. In this study, the electron beam energy was modulated to alternate between values just above the excitation energy of the 1s2s ^{3}S$_1$ level of interest and a value below, which would nevertheless maintain the same charge state balance. The x-ray signal of the 1s2s ^{3}S$_1$ decay proved to be a clean single exponential. In later experiments (for example, [44]), the beam was simply shut down completely after ion production and for the duration of the photon observation. This brings about the complication of a possible change of the charge state composition in the trap. In fact, when studying the radiative decay of an ion sample, it is necessary to monitor the number of ions in the ion cloud, some of which might get lost due to diffusion out of the observation volume or by charge exchange (or increase due to CX of more highly charged ions). Such monitoring is more straightforward without an electron beam that tries to restore the previous steady-state balance. If the vacuum is good enough, CX observations have shown an ion loss rate from the trapped ion sample that corresponds to storage time constants of many seconds. This ion loss is then a minor correction to the raw lifetime data of milli- or microsecond radiative lifetimes. The Livermore EBIT experiments have also demonstrated that it is essential not to overionize the sample, even as the signal rate may be increased that way—CX from higher charge states into the charge state of interest can spoil the accuracy of the lifetime measurement.

Atomic lifetime measurements on electric-dipole forbidden transitions in the visible (which are of particular interest in fusion plasma diagnostics and in solar and general astrophysics) were first demonstrated (among electron beam ion traps) at NIST Gaithersburg [9, 10], but with only moderate precision. Subsequent work at Oxford, Livermore and Heidelberg has investigated systematic error sources and drastically reduced their influence, so that the latest measurements (see below) challenge the reliability of modern theory.

At Livermore, atomic lifetime measurements of metastable levels in various ions by now range from

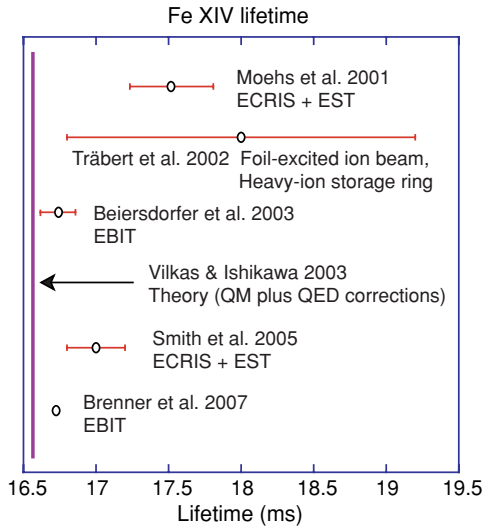

Figure 1. Timeline of lifetime measurements of the $3s^2 3p\ ^2P^o_{3/2}$ level in the ground term of the Al-like ion Fe^{13+}. The measurements are identified in the text. The vertical line on the left marks the lifetime expected from theory, with a QED correction to the M1 transition operator added. ECRIS: electron cyclotron resonance ion source; EST: electrostatic ion trap; EBIT: electron beam ion trap.

many nanoseconds [45] to many milliseconds [46]. The claimed measurement accuracy has reached 0.1% in several experiments at the Heidelberg EBIT [47, 48]. At this level of uncertainty, however, the results are at variance with complex quantum mechanical calculations that include QED not only in the energy levels, but also in the M1 transition operator. Such a discrepancy might indicate the presence of 'new physics', if the experimental errors, especially systematic errors, are truly understood at this level of accuracy.

A series of measurements [48–52] has targeted the magnetic dipole transition rate in the ground term of the Al-like ion Fe^{13+} (spectrum Fe XIV), which is also of astrophysical interest. The experimenters have used different types of ion traps (electrostatic ion trap, heavy-ion storage ring, electron beam ion trap) in order to determine a radiative level lifetime of about 16.6 ms, and the general trend of the measurements shows conversion towards the predicted value and shrinking error bars (figure 1). However, in this case (not in some others) all experimental results are on the long side of the expected lifetime result, which suggests the presence of a systematic error that is different from ion loss (which would make the apparent lifetime shorter). An experience from beam–foil spectroscopy we have is that too long lifetimes often are the result of cascade replenishment, but the conventional wisdom in highly charged ion trapping is that the technique addresses levels with lifetimes many orders of magnitude more long-lived than practically all higher-lying levels. Maybe to any rule there are exceptions?

For example, in figure 1 the heavy-ion storage ring measurement carries the largest error bar, although for many other atomic systems, the heavy-ion storage ring has yielded results of much smaller uncertainty and in agreement with data or trends of data from electron beam ion trap work. There is no readily apparent reason why these two types of ion traps would not both be delivering results of better quality that seems possible with electrostatic ion traps. Maybe we should read figure 1 not as a time line, but look at particle density or vacuum conditions. The electrostatic ion trap work was performed in a vacuum near 5×10^{-10} Torr; heavy-ion storage ring and (cryogenically cooled) electron beam ion trap operate with vacua that are an order of magnitude better. Neither vacuum can be blamed to cause the observed systematic shift. However, only in the electron beam ion trap are the ions are produced inside the same ultrahigh vacuum where the later lifetime measurement takes place; the experiments using electrostatic ion traps inject ions that have been produced in an electron cyclotron resonance ion source (ECRIS) which operates at high vacuum, but at a pressure that is several orders of magnitude higher than in the trap. Lastly, the heavy-ion storage ring in this case was provided with ions from an accelerator in which the ions were passed through a stripper foil (solid state density). Collisional–radiative modelling calculations suggest that non-selective excitation (as is taking place in the absence of specific measures) should result in the eventual population of one particular excited level ($3s3p3d\ ^4F^o_{9/2}$) that happens to feature a lifetime some 20% longer than that of the level of interest, while apparently all other levels are shorter lived by several orders of magnitude. The decay channels of this level converge on the ground term level of interest and replenish its level population. Such a small lifetime difference cannot be disentangled in the decay curve analysis. Cascade repopulation from this singularly long-lived level can then be understood to cause a significant systematic error, particularly badly for Al-like Fe ions, whereas for Al-like Ni^{15+} ions the lifetimes are different enough to be separable (for a discussion of the details, see [53]). The electron beam ion trap measurement should suffer the least from this cascade process, but possibly still more than the previous error estimates have allowed for. Apparently, among the three ion trap types employed for the data in figure 1 that measurement worked out best in which the ions were maintained at the lowest particle density all the time—that is inside an EBIT.

One motivation for such lifetime measurements is the intellectual quest of finding out whether quantum mechanics can be implemented well enough in the description of many-electron systems that in the combination with QED the atomic lifetime results are experimentally testable to high precision. At present, the aforementioned Heidelberg EBIT experiments claim an uncertainty of 0.1%, which is much more precise than the QED correction to the M1 transition operator (0.45%), and the experiment disagrees with good calculations on the order of 0.7%–1%. The special cascade may be estimated to reduce the discrepancy by up to about 0.3%, which would leave a notable mismatch with theory. Such a mismatch has not been seen in highly precise work on electric dipole transition probabilities in neutral atoms. If the discrepancies persisted and could be substantiated in improved experiments, they might indicate physics beyond the standard model. If then radiative lifetime calculations showed fundamental shortcomings, atomic structure calculations might also harbour surprises.

Another, more 'applied' interest is the benchmarking of extensive collisional–radiative codes that are used to describe

terrestrial and astrophysical plasmas. Here the requirement for measurement accuracy is not as extreme as for the fundamental physics context. Experimental uncertainties of the order of 1% are quite demanding, but have been reached in a number of measurements on trapped ions (see reviews [41, 42]), and such data should satisfy any foreseeable practical needs. The radiative–collisional modelling codes comprise thousands or ten thousands of levels and some ten times as many transitions. The energies of some levels may be known from measurements; the lifetimes of some low-lying levels with E1 decays may be roughly known from beam–foil or laser spectroscopic studies. Such detail almost vanishes in the complexity and richness of the models. However, the states in the ground configuration usually decay towards the true ground state by M1 or E2 transitions, with level lifetimes in the millisecond range. In low-density plasmas, these latter radiative decays compete with collisional excitation, with significant changes in the relative level population of those very low-lying levels as a function of density and a notable effect on certain line intensity ratios. Incidentally, these (few) lifetimes of low-lying levels can often be measured at a heavy-ion storage ring or in an EBIT, and thus a key element of the modelling effort can be validated. In fact, these lifetimes may be the best parameters that an experiment can study to test the modelling.

Most of the atomic lifetime measurements on highly charged ions concern M1 or M2 transitions. At the heavy-ion storage ring, some E2 transition rates have been measured as well. These studies complement the research into E1-forbidden decays that aim at ultraprecise atomic clocks using certain atoms or singly charged ions. At the Livermore EBIT, the highest multipole order decay studied has been a magnetic octupole decay (M3) as it occurs in Ni-like ions [54, 55]. Xenon, however, the element tried for lifetime measurements, has a number of isotopes and odd ones among them. Measurements on individual Xe isotopes have confirmed quantitatively a theoretical suggestion that the hyperfine interaction should mix E2 and M3 decays in this case [56], and thus certain hyperfine sublevels show a shorter lifetime.

4. Dynamic effects in plasmas

In spectroscopic observations of a plasma, one tries to keep the interrogation time shorter than the time scale of most variations of a plasma. This can be technically extremely demanding with short-lived high-energy density plasmas. Atomic level lifetimes always play a role in the variations. It would be advantageous to always have suitable atomic levels on which to study the mutual dependences and possibly on convenient time scales. The metastable levels of certain ion species have an influence on various balances in a plasma, for example the population distribution or the charge state distribution. Under steady-state conditions, this role may be modelled and inferred; if one perturbs the plasma towards a non-equilibrium state, the role of an individual level may be directly measurable. In a low-density plasma, metastable level lifetimes in the millisecond range can be very convenient because the radiative rates and the collision rates are comparable to each other.

I take as an example the $3d^9 4s\ ^3D_3$ level of Ni-like ions. This is the lowest excited level, and its only decay channel to the $3d^{10} J = 0$ ground state is by a magnetic octupole (M3) transition as mentioned above. When the charge state distribution in a hot Au plasma was modelled, there were clear discrepancies between expectations supported by HULLAC code radiative–collisional modelling and actual experimental data [57], until the role of the long-lived $3d^9 4s\ ^3D_3$ level in Ni-like ions was recognized. However, such a special role (see below) was explicitly denied for this level in the electron excitation calculations by Badnell *et al* [58] from which the level was intentionally excluded.

In a sequence of increasing charge state $q+$ of a given element, the ionization energy is a monotonically increasing function, defined as the energy necessary to remove a valence electron from one ion by exciting it to the ionization limit of the next higher charge state ion, which is in its ground state. For the Cu-, Ni- and Co-like ions of Xe this means ionization energy values of about 0.86, 1.50 and 1.58 keV. An electron beam of energy below 1.5 keV would not be expected to ionize the Ni-like ion Xe^{26+}, but an electron beam of not much more than this threshold value could ionize both Xe^{26+} and Xe^{27+} (Co-like) ions. Measurements at the Livermore EBIT have determined an excitation energy of 590 eV for the $3d^9 4s\ ^3D_3$ level of Ni-like Xe ions [59] and a radiative level lifetime of about 15 ms [55] (neglecting hyperfine effects). (Other levels of the same term have nanosecond lifetimes, while most other levels of the same ion have lifetimes in the picosecond range.) A Ni-like ion in this excited state can be ionized by another electronic excitation of at least 910 eV. This means, the metastable level provides a stepping stone, and the effective ionization threshold is lowered from 1.50 keV to merely 0.91 keV. This is more than a curiosity on paper: in observations at Livermore, the (Co-like) spectrum of Xe^{27+} [60] was indeed produced [61] with an electron beam energy well below the 1.5 keV value of the standard definition of an ionization potential (figure 2).

When the electron beam is switched on at the beginning of a trapping cycle, the ions of the various charge states are sequentially produced. By the energy argument, the Co-like ion should be produced almost as soon as there are any Ni-like ions, and they are seen in the spectrum. However, an observed time delay seems to indicate that the ionization towards the Co-like ion requires notably more time—which may be interpreted as evidence for ionization through a multistep pathway of limited phase space. (We are working on a simulation of the time dependence of the spectra [61].) Hence individual atomic levels can have a notable influence on the rate with which a plasma adjusts to changes of external conditions.

In the above example, the collision time is shorter than the level lifetime, so this is not a very dilute 'coronal' plasma, and the effect described happens in most technical plasmas. Basically all noble gas-like ions provide such stepping stones, and care has to be taken if one wants to avoid reaching the next higher charge state. Spectra have been misinterpreted and excitation cross sections misrepresented, because the

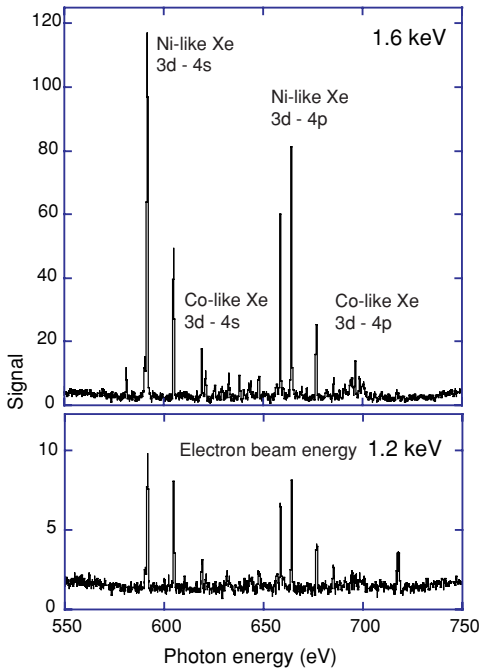

Figure 2. Lines typical for Xe XXVIII (Co-like Xe^{27+} ions) are seen to be produced at an electron beam energy of 1.2 keV (lower graph), well below the 1.5 keV ionization potential of Xe XXVII (Ni-like Xe^{26+} ions). Electron beam energies of 1.6 keV (upper graph) and higher are sufficient to ionize Ni-like as well as Co-like ions of Xe.

contamination by a higher charge state was overlooked. It has also happened that collisional–radiative modelling predicted unrealistically high charge states, because the lifetime of such metastable levels was not put at its finite value, but was implied to be infinite.

5. Outlook

Electron beam ion traps have had a remarkable career as sources of ions that could either be kept trapped (and studied) or ejected as a beam in order to serve collision studies outside the trap (after all, EBITs started out as a derivative of earlier electron beam ion source (EBIS) designs). The extracted ion beams can also be analysed in terms of the charge state distribution inside the device. (Examples from various EBIT laboratories are [17–19, 62–64].) The capabilities of the device, however, are far from exhausted by the examples discussed above. Owing to the low density of the ion cloud in an EBIT, this certainly is not a particularly bright light source, but it is enormously versatile. For example, recent applications have used an EBIT as a multi-line x-ray source for measurements of x-ray filter transmission data [65] for the benefit of laser-driven fusion experiments. The internal dynamics of the trapped ion cloud has been studied on and off in many ways over the years. Investigating the ion plasma response to perturbations remains an interesting field.

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